LIMESTONE DISSOLUTION PROCESSES IN BEKE DOLINE AGGTELEK NATIONAL PARK, HUNGARY

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ABSTRACT

Aggtelek National Park, Hungary, is a limestone karst upland characterized by karren, dolines and river caves. For a period of two years, climatic and carbonate dissolution variables were monitored at four depths in a 7·5 m shaft through the soil fill in the floor of a typical large (150 m diameter) doline. Results are compared to other monitoring stations in shallow soils on side slopes. Runoff and groundwater flow are focused into the base of the doline soil fill, where moisture is maintained at 70–90 per cent field capacity and temperatures permit year-round production of soil CO_2. The capacity to dissolve calcite (limestone) ranges from *c.* 3 g m^{-2} per year beneath thin soils on the driest slopes to 17–30 g m^{-2} per year in the top 1–2 m of doline fill and at its base 5–7 m below. © 1997 by John Wiley & Sons, Ltd.

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INTRODUCTION

Dolines or sinkholes are diagnostic features of karst terrains. They are topographically closed depressions with forms that may range from shaft through funnel to shallow bowl. Diameters range from less than 10 m to a little more than 1000 m, and depth:diameter ratios are normally between 1:2 and 1:20. Processes of mechanical collapse, subsidence or suffosion may contribute to their formation but in true karst (as opposed to pseudo-karst) aqueous dissolution of the bedrock is either the principal process or the essential trigger for operation of any of the others. In karst areas where limestone or dolomite outcrop, dissolution tends to be the quantitatively predominant process (Ford and Williams, 1989, pp. 396–405.

In forest or grassland regions, dissolution dolines typically are mantled with soil. There is often some outcropping of bedrock displaying karren forms on the steeper sideslopes, and a marked increase of soil thickness in the bottoms. The focusing form of the doline funnel, with its potential for retaining greater moisture and perhaps generating higher PCO_2 in soil in the base, has led to much speculative discussion concerning positive feedback effects that deepen adjoining dolines and thus expand their diameters until their perimeters come to overlap, creating a polygonal karst (Jakucs, 1977; Williams, 1985).

In many karst areas, human-induced soil erosion has greatly increased the exposure of karren on doline side slopes and the detrital infilling of their floors. In extreme cases their basal drains become blocked (deliberately or inadvertently), thus terminating their hydrogeological function; temporary or permanent ponds are created, as reported below.

Despite the interest in dolines there have been few attempts at modern, instrumented study of the dissolution process at work in them. Limestone and dolomite dissolution rates under soil are determined primarily by the availability of CO_2 and water. In the general pedological literature there is much discussion of soil CO_2 production and dispersion (e.g. Monteith *et al.*, 1964; Matthess, 1973; Trainer and Heath, 1976). It focuses on the A and B horizons and rooting zone, with sampling rarely extending below 0·5 m. In humid mid-latitude sites, the general finding is that soil moisture is at a maximum during the winter or the spring thaw; there is a

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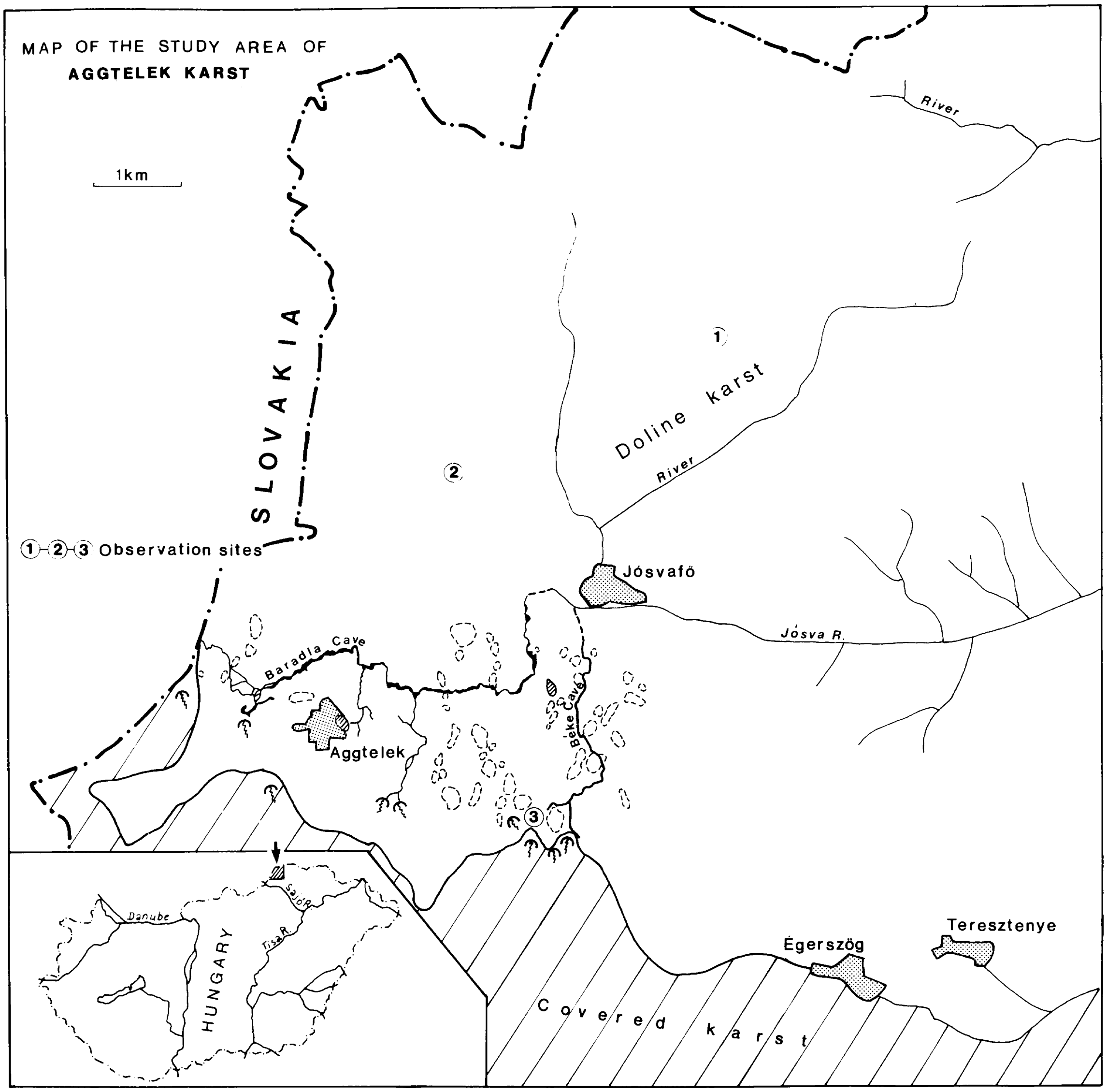

Figure 1. Location of the Aggtelek Karst, Hungary. The principal field site reported in this paper is Site 3 – Beke Doline

seasonal bloom of CO_2 with a maximum in the late spring or summer. All authors note that there is much intra- and inter-site variation. One hydrogeological study in Canada that extended to greater depths (to a water-table at 7–11 m in glacial outwash sands) demonstrated the progressive seasonal bloom particularly clearly (Reardon *et al.*, 1979). There have also been many studies of soil CO_2 and associated carbonate dissolution in different climates by karst geomorphologists since the pioneering work of Miotke (1972, 1974). Once again these have been limited largely to sampling in the upper 50 cm or so of hillslope soils or depression fillings (e.g. Brook and Ford, 1982; Gunn and Trudgill, 1982; Crowther, 1983, 1984).

The purpose of this paper is to summarize some of the principal findings of what we believe to be the first study of limestone dissolution throughout the column of clastic filling in a large doline that is typical of those in the temperate, but not glaciated, regions. It investigates the positive feedback hypothesis. The work was carried out by the first author and is part of a larger, ongoing study of karst processes and karstic soils in the region

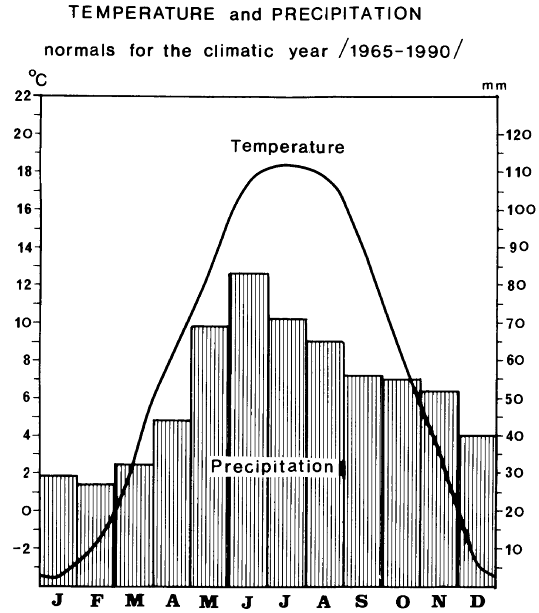

Figure 2. Thirty-year mean monthly temperature and precipitation records for the Aggtelek region, Hungary

concerned. Much has been published already in Hungarian sources (Zambo 1971a,b, 1973, 1985, 1993a–d), the intent here is to review the results and expose them to a wider readership.

THE STUDY AREA–AGGTELEK NATIONAL PARK

The study site is located in Aggtelek National Park, a rolling upland karst that straddles the Hungarian–Slovak border (Figure 1). Elevation ranges from 200 to 600 m a.s.l. Bedrocks are thick to massively bedded platformal limestones and dolomites of Triassic age. Insoluble residua are only 0·1 to 0·35 per cent (Zambo, 1971a). There was quite intensive karstification during the Cretaceous, after which the strata were buried by Pannonian sands and clays (Miocene). They were uplifted as a horst block in the Pliocene, when the modern period of erosion began. They display dense tectonic fracturing. In combination with the palaeokarstification, this has resulted in an unusually high density of initial fissures that are penetrable by groundwaters.

Today the area is comprehensively karstic. All observed bedrock surfaces, exposed or soil-mantled, are carved into karren forms. There is some buried pinnacle karst. Dolines are abundant, forming polygonal karst except on the highest ground and on some slopes. Their dimensions range from 30 to 300 m in diameter and they are up to 20 m in depth. A series of streams that collect on the Pannonian strata sink at the contact with the limestone and have generated the remarkable Baradla System and other river caves within it (Figure 1). The caves are of the ideal water-table type (Ford and Williams, 1989, p. 262), which implies that there is unusually high effective fissuration below the epikarst zone in these rocks.

Soils developed on the carbonate rocks are terra rossas and rendzinas (Kubiena (1950) red to red-brown chromic luvisols and rendols in the terminology of the *Seventh Approximation* (1960)). They have an effective porosity of 30–50 per cent (Bárány, 1980; Zambo, 1986). On slopes they are generally mixed and reworked to form a complex mosaic of relict and modern components. In most cases they are less than 50 cm in thickness where the slope is greater than *c.*16°, and may be absent entirely. A mediaeval vineyard was probably responsible for particularly severe erosion on a SW-facing slope that overlooks the village of Aggtelek (Kordos, 1975); the derived soil accumulated in a large doline at the contact with Pannonian clays, converting it into a permanent pond (Figure 1), despite the fact that a large, dry passage of Baradla Cave passes only 30 m beneath its floor.

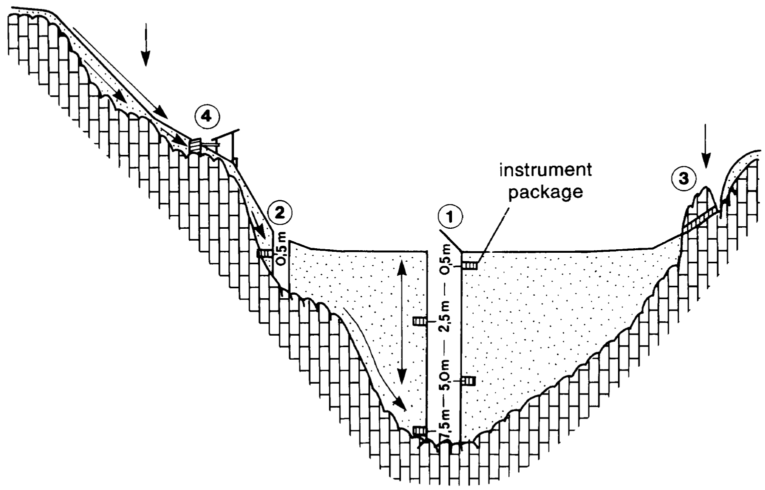

Figure 3. Cross-section of Beke Doline to display its modern instrumentation. There is vertical exaggeration of the scale for the sake of clarity. Results for Station 1, the doline base, are considered in this paper. Stations 2–4 are later additions

The natural vegetation of the region consisted of oak (*Quercus petrae*) and hornbeam (*Carpinus betulus*) forests with lesser beech (*Fagus* spp.), linden (*Tilia* spp.) and ash (*Fraxinus* spp.) on the steepest slopes. As the area is now a protected park, these species have returned and are mixed with plantations of spruce and pine, and grasslands. The experimental doline described here is occupied by the oaks plus aspen (*Populus tremula*), juniper (*Juniper communis*) and blackthorn (*Prunus spinnsa*) bushes.

The 30-year temperature and precipitation means for major meteorological stations in this region are displayed in Figure 2. Mean precipitation is *c.* 560 mm a^{-1}. Topoclimatic factors may cause considerable deviations from these values locally (e.g. Novaky, 1991), and there is substantial year-to-year variation at a given site. It was established in this research that the amount of rain falling in an event is particularly significant in the context of subsoil dissolution. A minimum of 5 mm precipitation was necessary to produce dissolution beneath even the thinnest soils of the area. There were 125 rain days during the two years of the study, but the 5 mm value was exceeded in only 33 of them. Rainfall intensity of 0·1 mm min^{-1} or greater was necessary to generate surface runoff.

Net snowfall amounts to 40–140 mm water equivalent per year, with much of this variation attributable to local orography and drifting. The slow infiltration of snowmelt can be more efficient in achieving dissolution than many rain storms that supply equivalent amounts of water. Mean annual actual evapotranspiration is estimated to be less than 400 mm on the karst.

THE EXPERIMENTAL INSTALLATIONS AND MEASUREMENTS

Four dolines at three different sites were selected for study (Figure 1) but the comprehensive instrumentation had to be limited to one site. This is a nearly circular feature, 150 m in diameter and 12 m in depth. Its basal fill of soil is a further 7·5 m in depth (Figure 3). The doline is situated over Beke Cave, which is 37 m beneath it. There are now four different measuring stations or experiments operating in the doline, plus a further one in the cave below. However, only the doline bottom station (Station 1 of Figure 3), was operating during the measuring years considered here; its data will be compared with data from a sample of two of eight shallow subsoil stations that were also operated in those years. These latter stations were installed at the soil–rock interface in typical doline sideslopes where the soil thickness was less than 50 cm. They were distributed to investigate any effects of differing slope gradient and aspect. Their instrumentation was the same as in the doline.

In the doline, a square access shaft measuring 1·5 m on each side was carefully dug by hand to the bedrock floor at −7·5 m. The infilling materials are silts and clays. Quartz is the principal mineral, but plagioclase, gibbsite and pyrite are prominent at some stratigraphic levels. Clay minerals (chiefly illite and lesser kaolinite)

represent 9–30 per cent and organics 1·8–6·5 per cent. The filling is divided into 31 successive layers that are easily recognizable to the naked eye: their differing colours (from red: 2·5 YR3/4 to dark brown: 10 YR2/1) are created by varying abundances of Fe, Al and Mn oxides and hydroxides (Zambo, 1985, 1986). Filling possibly commenced at the close of the Miocene. The accumulation below −5 m is attributed to the Pliocene, and that between −0·7 and −5 m to cooler Pleistocene conditions (Zambo, 1993a,b). The uppermost 0·7 m of fill probably resulted from human activity. The bedrocks at the base of the shaft have a rounded karren form and are fissured, draining readily.

After digging, the shaft walls were sealed with impermeable sheeting buttressed by timber to restore the natural drainage of the undisturbed soil infill. It is considered that this was successful (Zambo, 1986). Four instrument packages were inserted, one in each wall, at depths of 0·5, 2·5, 5·0 and 7·5 m; their electronic output was linked to a Sharp computer housed outside the doline. Each package comprised an open-topped aluminium box, of dimensions 100×80×15 cm, that was inserted into an accurately dug lateral cavity in the soil. The box contained a plastic water-collecting tray, of dimensions 60×40×5 cm, that was filled with polypropylene balls to serve as a mechanical support and inert filter, and jacked against the cavity roof (i.e. the local soil base). The outlet from the box to the shaft was sealed airtight and watertight. Water collecting in the trays drained gravitationally into sealed bottles in the shaft, where its rate of accumulation could be measured continuously and it could be sampled periodically for analysis. Regular control measurements established that soil moisture drainage into the trays was not significantly faster or slower than that elsewhere in the doline fill, and that the partial pressure of CO_2 in the collecting chamber was the same as that elsewhere at that depth in undisturbed soil.

Soil temperature was measured with Wille 400-type thermistors inserted to depths of 60 cm alongside the collecting chambers, plus standard glass thermometers to check their calibration. Wille-type soil moisture analysers (conductivity bridges) were used to determine soil water content. CO_2 was measured by Draeger tubes and a pump withdrawing from the sealed collecting chamber, with an accuracy of 0·01 per cent. Field pH was measured with a portable pH meter.

For comparison, measurements with the same instrumentation and installation at two sideslope sites are given. Site 4 is on a 25° slope with a northeast exposure and soil depth of 0·2–0·45 m. Site 7 has an eastern exposure, 20° slope and 0·2–0·5 m of soil.

For the results reported here, the water-collecting bottles were sampled and CO_2 measured every 14th day, plus special visits following exceptional rains. A total of 53 collections were made during the two-year period, May 1981–April 1983. At each station at each collection, three 50 ml bottles were filled and sealed for laboratory analysis of (i) total CO_2, (ii) complexed CO_2, and (iii) pH and the common cations Ca, Mg, Si, K, Na, P and Ti as oxides.

RESULTS AND DISCUSSION

Ground temperatures

All of the steps involved in the production of CO_2 and carbonic acid in the soil, and of its dissolution of carbonate rock there and below, are temperature-dependent to some extent (Drake, 1984). The temperature records of the doline station and of the two selected sideslope sites are given in Figure 4. Results are largely as expected. The amplitude of the annual cycle in the thin soils is 12–15°C, with the actual temperatures being close to 0°C for three to four months each winter; they fell below this value at Site 7 in the cold winter of 1981–2. These low temperatures will bring the biological production of CO_2 to a halt, significantly reducing the solvent potential of any waters passing through them at the time.

In the doline, the amplitude of the annual cycle is reduced to about 8°C at −2·5 m, 4° at the −5 m level and 3° at the base. A most interesting feature of the records is that the base is slightly cooler than at −5 m. This reflects the effects of occasional influxes of cold water from the doline sides, where it is evidently flowing preferentially at the soil–rock interface. They are marked by some quite pronounced cooling events with comparatively rapid rates of change, e.g. November 1981, January 1982 and February 1983. Nothing similar is recorded at −5 m.

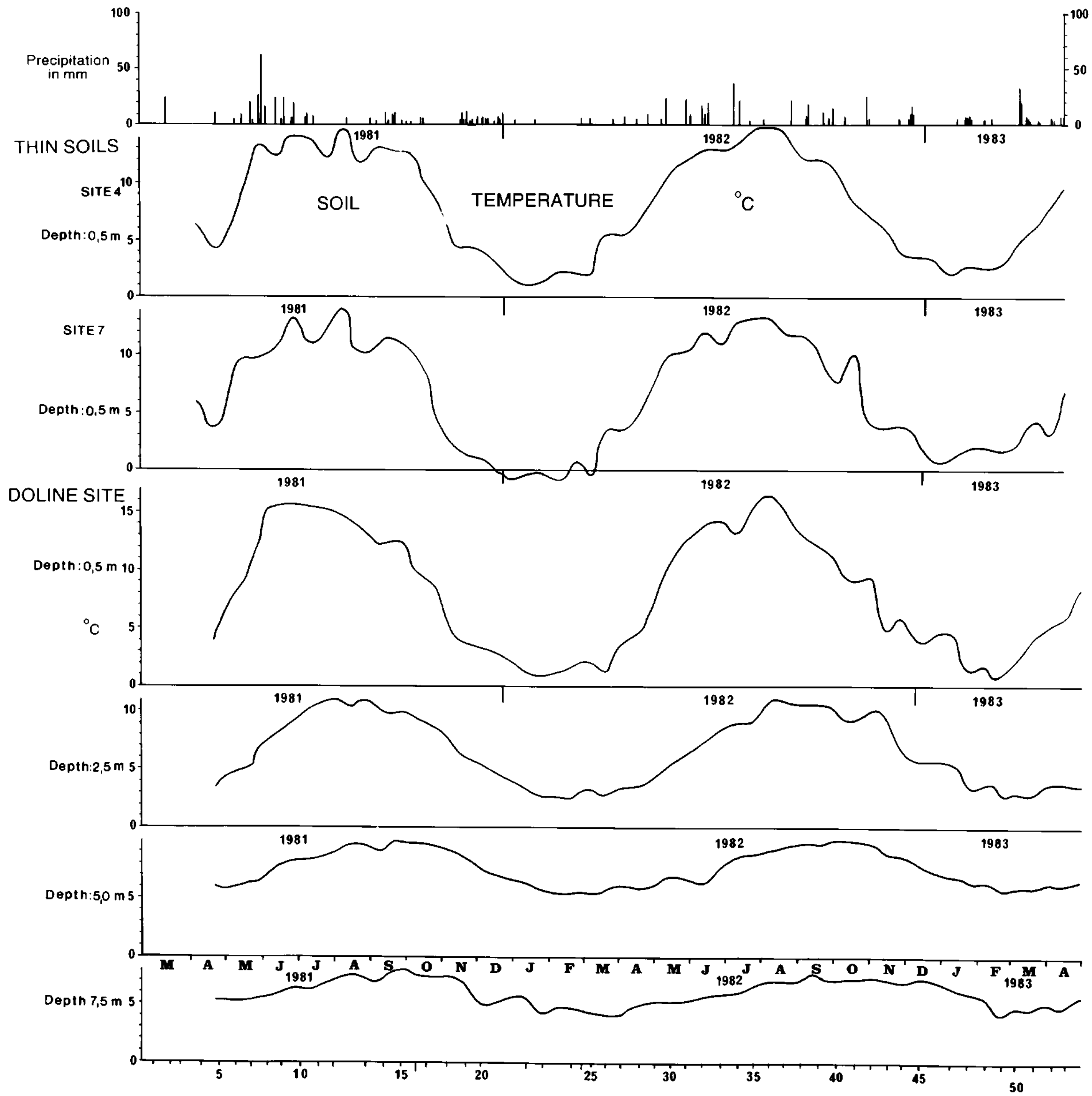

Figure 4. Precipitation and soil temperature records for Beke Doline (Station 1) and two sideslope sites (Sites 4 and 7), March 1981–April 1983

The most significant features of the graphs are that bacterial production of CO_2 may be expected to continue throughout the year below 2 m or more of filling in a doline in these climatic conditions, and that CO_2 solubility will be somewhat enhanced in the cool environment of the doline base. The most important rate control of carbonate dissolution then becomes the supply of water, and the temperature records provide a first hint that this may also be enhanced close to the doline base.

Infiltration and the soil moisture regime

Soil water flow recorded in the collecting trays and the soil moisture conditions estimated by conductivity are shown in Figures 5 and 6. In the experiments reported here, the eight sampling stations in thin soils were distributed to investigate the possible effects of differing slope gradient, slope aspect and form (convex or

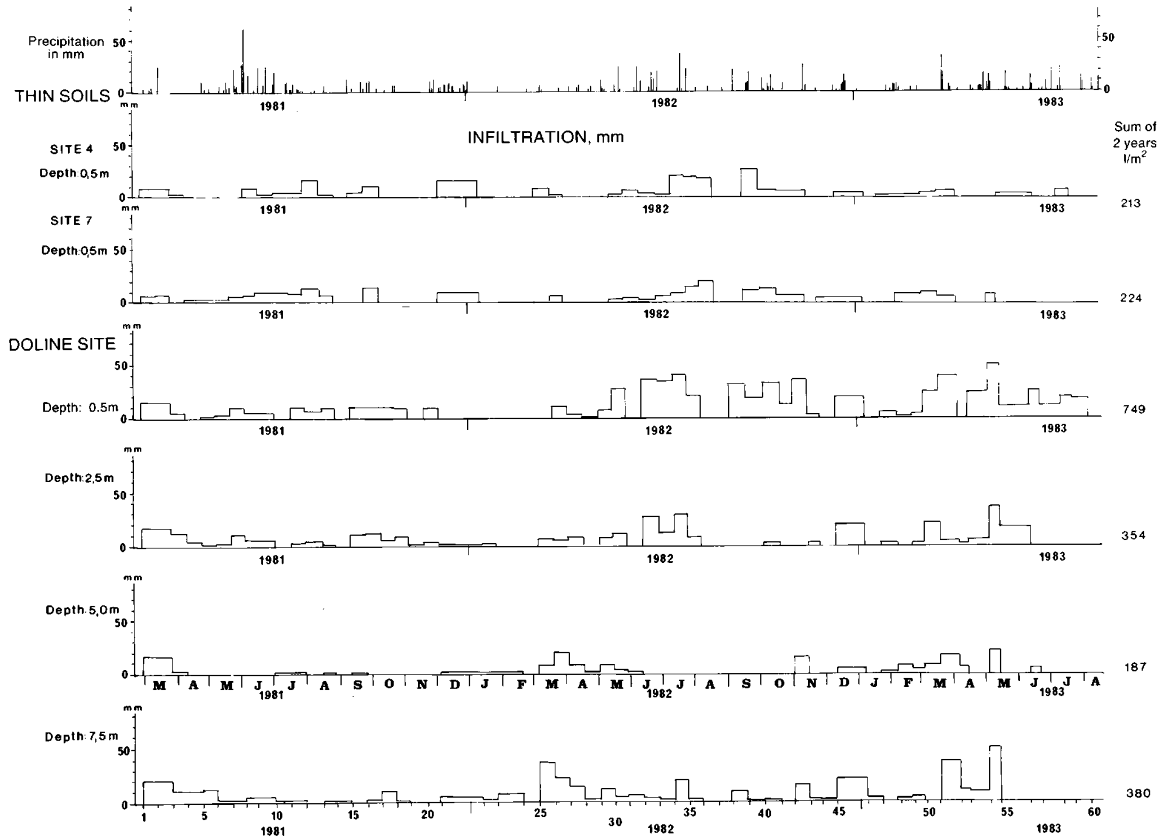

Figure 5. Infiltration measured at Beke Doline (Station 1) and two sideslope sites (Sites 4 and 7), March 1981–April 1983

concave). It was established that infiltration decreases as gradient increases, as many other workers have found; here, only 14 per cent of measured annual precipitation was intercepted on slopes steeper than 30°, increasing to more than 21 per cent on slopes below 15°. The nearly flat surface at the centre of the doline was able to adsorb 37 per cent of the precipitation.

Most of the recorded soil water flow is correlated with particular precipitation events (Figure 5). The early summer peak of precipitation that occurs in this region supplied 40–50 per cent of the annual infiltration that was recorded at each thin soil station, regardless of its gradient or aspect. A secondary precipitation maximum in the autumn was marked by some differentiation: east-facing slopes conserved significantly more water as infiltration.

Figure 5 records some distinctive features in the doline. The lower permeability of the middle layers of its infilling evidently inhibit infiltration in the centre of the doline. At −5 m this was only 13 per cent of the precipitation, compared to 37 per cent at the surface. The deficit represents evaporation losses, which are enhanced by the early summer maximum of infiltration water supply. Infiltration was most continuous at the base, although the coefficient of variation there remains high. There are strong winter and early spring peaks. A surge of groundwater at and below −2·5 m in March 1982 is attributed to thawing snow.

In Figure 6 soil moisture conditions are estimated as a percentage of the field capacity at the sample stations. Porosity of the soils ranged from 30 to 50 per cent, variations being chiefly due to differing grain sizes and to the accumulation of cutans or Fe and Mn oxides. It is seen that truly saturated conditions were not recorded by the measuring techniques used here. The thin soils maintained values close to 60 per cent for much of the year but displayed late summer droughts when their temperatures (and thus their potential CO_2 productivity) were at their greatest. The sharp drop recorded in January–February 1982 at Site 7 was due to the prolonged freeze-up

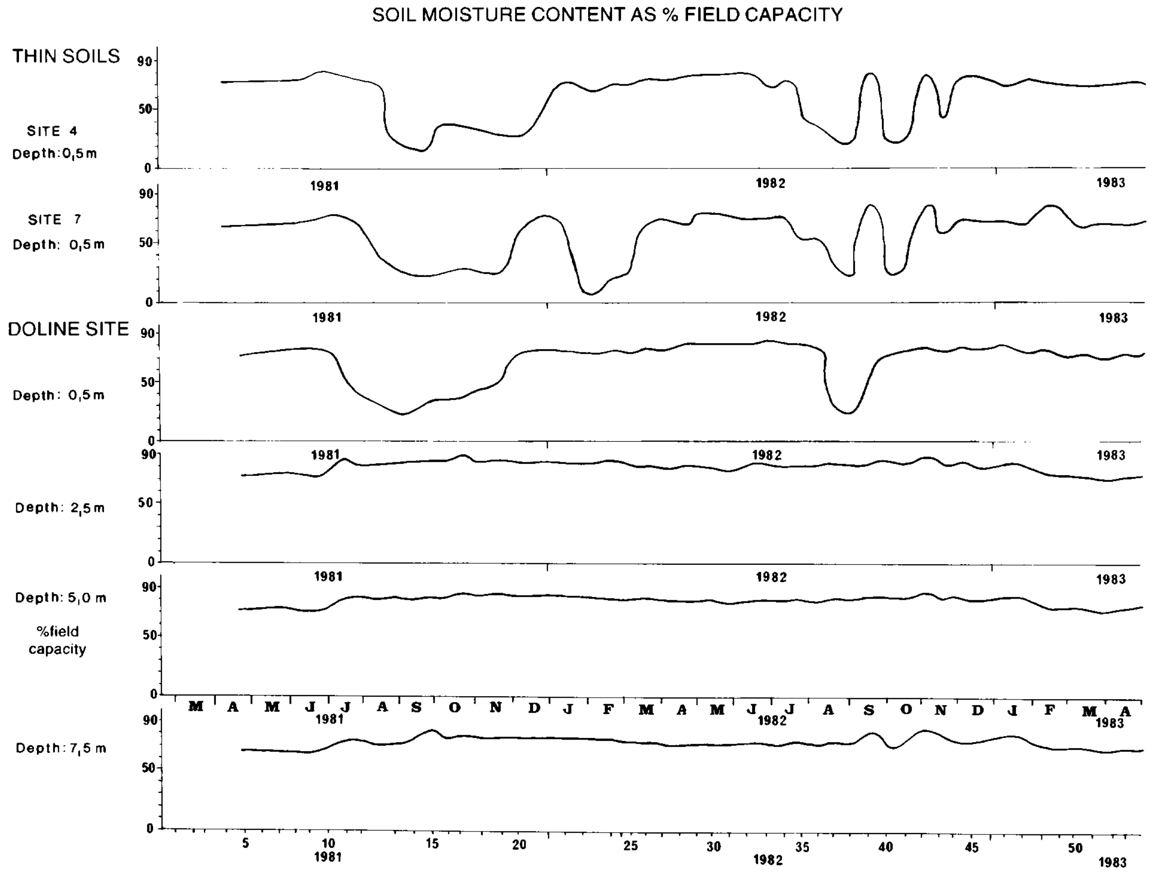

Figure 6. Soil moisture estimated as percentage field capacity at Beke Doline (Station 1) and two sideslope sites (Sites 4 and 7), March 1981–April 1983

that occurred there (Figure 4). The 0·5 m station in the doline displays a similar record although, owing to insulating snow, there was no freezing. The –2·5 and –5 m stations in the middle, less permeable, layers of the infilling were nearly constant at 70–80 per cent capacity, comparable to the results that Reardon *et al.* (1979) obtained at similar depths in outwash sands. This must be judged more than sufficient to maintain bacterial production of CO_2 in the supportive thermal environment at Aggtelek. There is a little greater fluctuation of moisture content at the base of the doline, associated with the lateral influxes of water there, but it also remains at 70 per cent or higher, which is somewhat more than might be predicted given the opportunities for vertical drainage downwards into the fissured limestone immediately beneath.

Carbon dioxide in the soil air

As noted, the void ratio in the soils and deeper fill of the doline ranges between 30 and 50 per cent. On average, the thin soils stations and the –0·5 m station in the doline contained 9 volume per cent of air, increasing to as much as 20 per cent during droughts. The mean value was reduced to 6–7·5 volume per cent in the deeper stations, with no significant drought effect. When saturated with water it is estimated that 2–3 volume per cent of air will be retained.

These proportions of air will permit oxidation and microbial fixing processes to consume 0_2 quite rapidly in the temperature conditions that have been defined. The CO_2 content rises. In the shallow soils and the upper levels of the doline, plant root respiration and aerobic bacteria are believed to be the principal contributors. In the deeper stations there is probably continuous production of CO_2 by fermentation and anaerobic bacteria, plus strong seasonal blooms attributable to aerobic microflora. Aeration is considered to be comparatively poor in

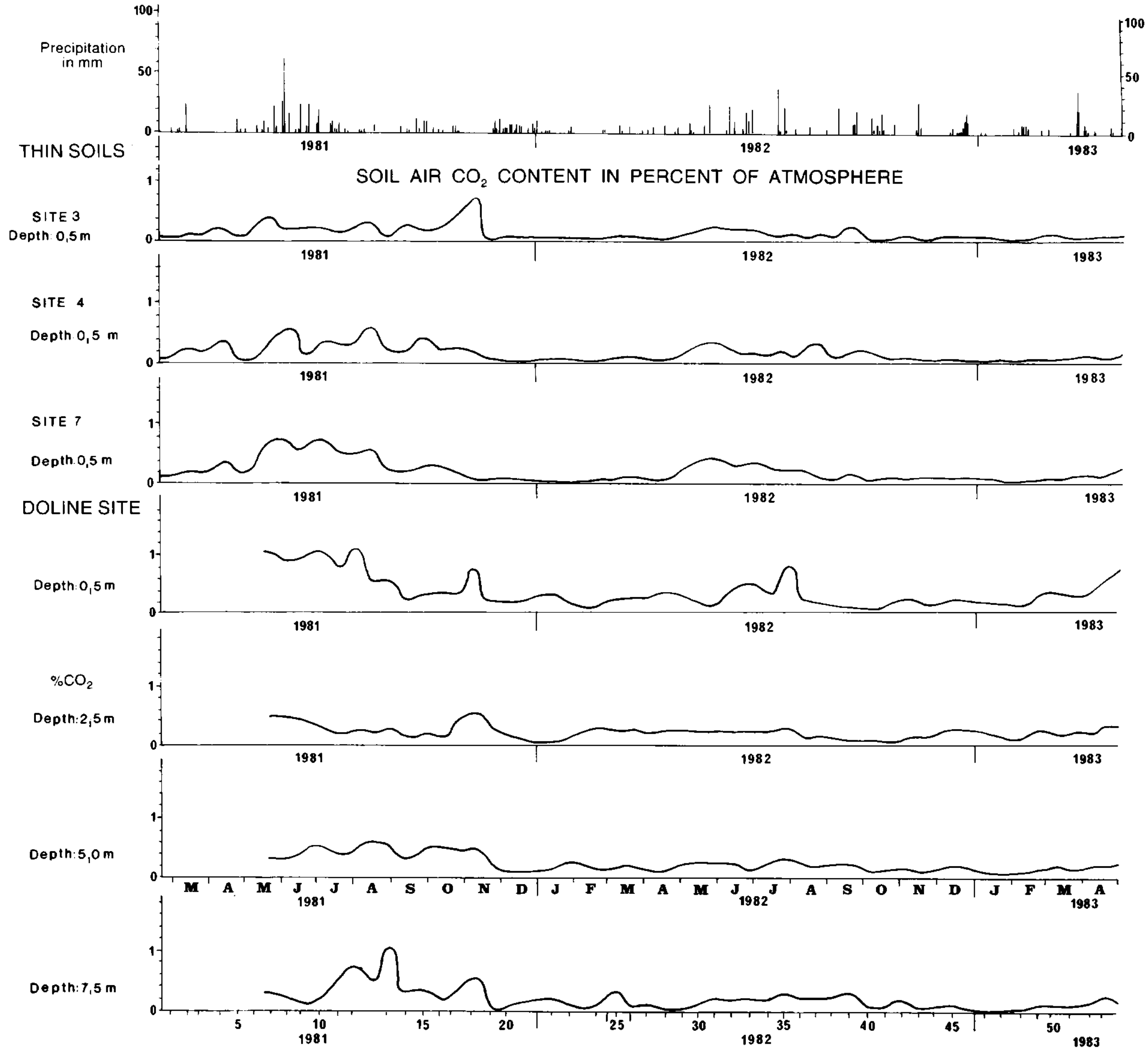

Figure 7. CO_2 as percentage concentration in soil air at Beke Doline (Station 1), and two sideslope sites (Sites 4 and 7), March 1981–April 1983

terra rossa soils (Zambo, 1986), but the results discussed here suggest that it is sufficient to sustain oxidation processes at the stations for most of the time.

The two-year mean PCO_2 from all stations (889 measurements) was 0·43 per cent, but the mean of the thin soils alone was only half of this value except where the profile was unusually rich in humus. These data should be compared with the standard atmospheric mean PCO_2 at sea level of about 0·035 per cent. The ranges and mean values of the individual stations are a little below those reported from the shorter period, shallow depth (≤60 cm) studies in other extraglacial karsts in Malaysia (Crowther, 1984) and New Zealand (Gunn and Trudgill, 1982).

The temporal record is displayed in Figure 7. There are strong seasonal and year-to-year contrasts. To generalize, in a climatic year a first peak of CO_2 production tends to come 10–15 days after the beginning of snowmelt. It then falls in later spring before rising to an annual maximum which, at different stations and in different conditions, can occur at any time between late May and the end of August. Winter PCO_2 is less than

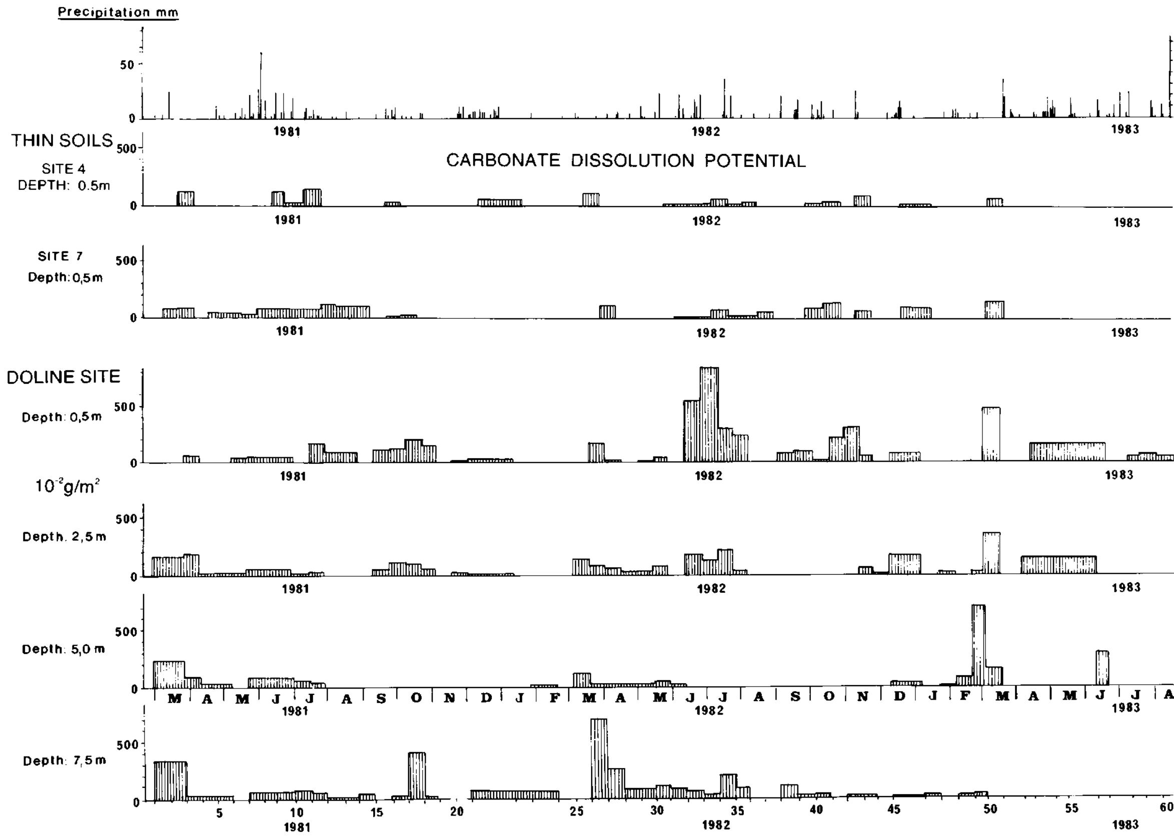

Figure 8. Carbonate dissolution potential at Beke Doline (Station 1) and two sideslope sites (Sites 4 and 7), March 1981–April 1983. The potential is calculated for pure $CaCO_3$ dissolved in thermodynamic equilibrium with the ambient temperatures and partial pressures of CO_2 that were recorded

one-third of the summer mean value in the shallow soils, as Miotke (1972) also found. This difference is reduced at depth; PCO_2 did not fall below 0·2 per cent at the –2·5 and –5 m stations. Productivity was also stable at the base of the doline during winter, but actual PCO_2 values measured there were somewhat lower, which can be attributed to extraction in the water flowing in from its sides.

The strong contrast between summer and autumn values recorded at all stations in 1981 and those of the later years must be viewed with caution; problems of reliable supply compelled a change of manufacturer of CO_2 sensor tubes in 1982. However, total early summer (mid-May to mid-July) precipitation was 280 mm in 1981, 186 mm in 1982 and 206 mm in 1983. These greater rains are believed to explain at least a part of the higher soil CO_2 measured in 1981. These results emphasize the need for multi-year series of measurements of such variables; multi-year records have not often been published in the karst literature.

Contrasts within the doline are of primary interest. There is the expected damping of CO_2 fluctuations at –2·5 and –5 m when compared to –0·5 m, but this effect was not so strong at –7·5 m. It is tempting to correlate a 1 per cent CO_2 peak occurring at mid-July 1981 at –0·5 m with one of late August at –7·5 m. However, it must be noted that all four levels in the doline registered a sharp peak of 0·4–0·6 per cent early in November that year. These data do not yield simple correlations when they are subject to serial analysis, nor is there good correlation with specific antecedent rainfall events.

Carbonate dissolution potential

Carbonate dissolution potential (Figure 8) is taken to be the amount of pure calcite ($CaCO_3$) that can be supported in solution at thermodynamic equilibrium in the prevailing conditions. As noted, the limestones of the region yield little insoluble residue, so that their bulk solubility will be similar to the potential value.

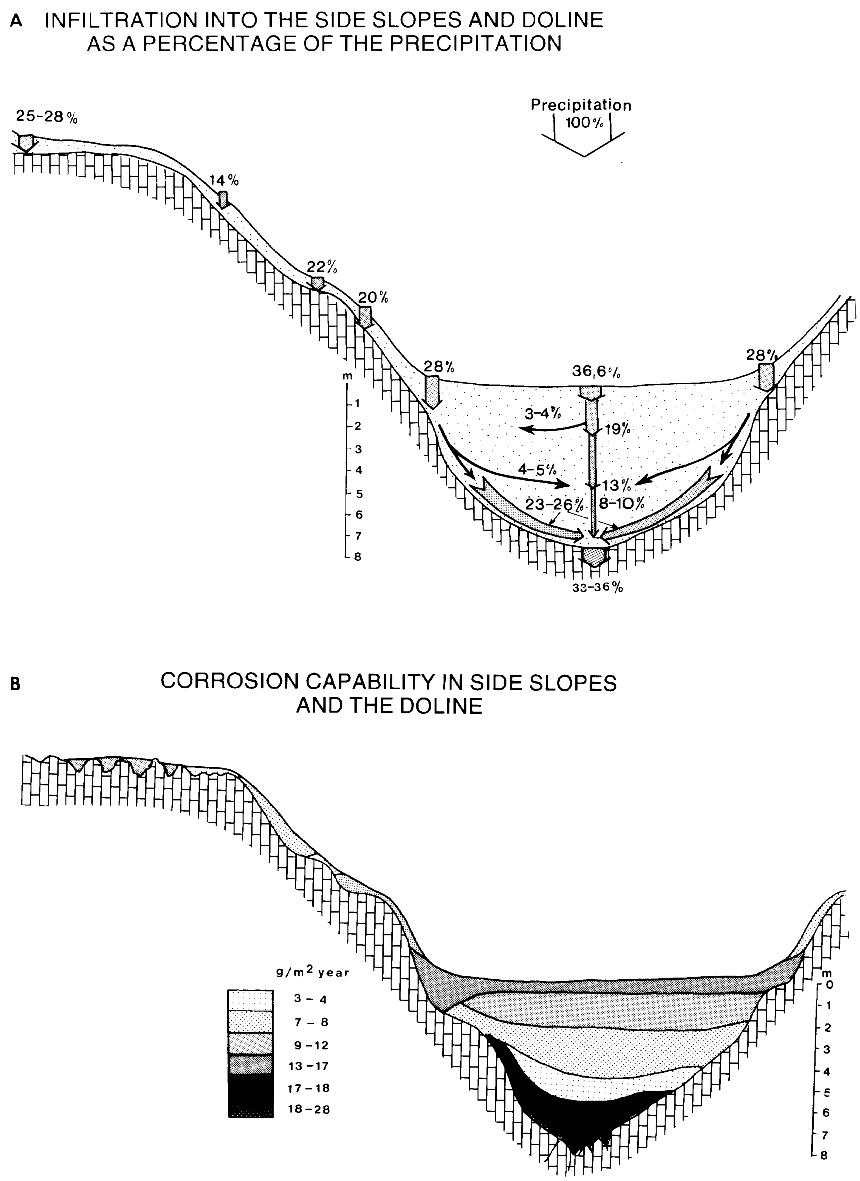

Figure 9. (A) Doline and sideslope groundwater infiltration expressed as a percentage of precipitation recorded at Beke Doline, the two sideslope sites and two other dolines, March 1981–April 1983. (B) Mean annual $CaCO_3$ dissolution rates for the doline and sideslopes calculated from the data presented above.

The mean concentration of 'free' CO_2 (i.e. that which is available to react with calcite) in rainwater was found to be 0·28 mmol l^{-1}. The mean for soil waters was 2·09 mmol l^{-1}, an enhancement of 7·5 times. For the mean annual flux of infiltration water this yields a solvent capacity of 26·65 g m^{-2} per year, or *c.* 10 000 m^3 km^{-2} per 1000 years if expressed in Bubnov units (e.g. Atkinson and Smith, 1976). From the solute loading data, some 9 g of this capacity is exhausted in the terra rossa and rendzina soils in the average case. In principle, the capacity to corrode fresh bedrock is therefore approximately 17 g m^{-2} per year. The mean capacity for 379 water samples taken at the soil–rock interface or below it was computed to be 12·7 g m^{-2} per year at equilibrium. The difference of some 4 g in these different estimates of capacity can be attributed to sampling and measurement errors, to the complexing of CO_2 with other ions, and to de-gassing losses.

The substantial spatial variations that were discovered within the regional mean data are displayed in schematic form in Figure 9. Given the minor temperature constraints that have been noted, the quantity of infiltration water is the rate control at any given location (Figure 9A). This is combined with the calculated solvent capacity (adjusted for the 4 g analytical differential) to yield the potential rate model of Figure 9B. This should be considered to be preliminary in many respects but it does emphasize a 'positive feedback' characteristic and other features that we believe to be valid and novel. The following are examples.

(i) Steeper slopes above the level of soil filling in a doline or in inter-doline terrain will tend to be self-sustaining where subject to dissolution only, because of their low potential dissolution.

(ii) In contrast, there is exceptionally high potential for dissolution in the top 0·5–2·0 m of any soil infill; if that fill is maintained at a constant elevation in a doline, a corrosion bank or terrace can be expected to form around its perimeter. Shallow, rough terraces that occupy one or a few portions of the perimeter only are often exposed as a consequence of intensive cultivation in dolines of central and southern Europe and southeast Asia.

(iii) The middle zones of doline fill tend to display lower porosity and permeability because of compaction and illuviation (with greater complexity where human activity has accelerated soil erosion) unless they have been disturbed by piping or collapse of the basal fill. The zones may become quite impermeable and conserve clasts from the side slopes; large, rounded limestone blocks often occur within them. Where these conditions extend to the doline floor the bedrock is protected from dissolution, and a pond may accumulate on the surface.

(iv) The basal fill (generally below 5–8 m in this region) usually has greater porosity owing to dissolution of the underlying bedrock. The corrosion potential is high to maximal if flow at the soil–rock interface can be focused there: rates of 18–30 g m^{-2} per year have been calculated. Beke Doline is a particularly good test case for the focusing effect because (as has been emphasized) the limestone possesses an unusually high frequency of penetrable fissures that are available to intercept groundwater flow before it reaches the doline bottom. Despite this factor, it has been demonstrated that significant basal enhancement of flow and of potential dissolution is a reality.

The gross limestone dissolution rates obtained in this study are a little below those of most of the other karst studies cited above, but are of the same order of magnitude. The extrapolated mean rate of lowering of the land surface is about 0·5 cm per 1000 years. On the inter-doline crests and the steeper sideslopes this reduces to 0·4 cm per 1000 years or below, while at the base of the doline filling it is increased to 0·7–1·0 cm per thousand years. The differential deepening rate obtained for the doline is thus in the range of 0·3–0·6 cm per 1000 years or about 3–6 m per million years. The difference in elevation between the base of its filling and the lowest points on its perimeter today is some 20 m: this suggests a differential dissolution age of 3·5–7·0 million years, which is in good agreement with the late Miocene/Pliocene age for the initiation that is suggested by features of the stratigraphy of the clastic fill.

The measuring programme at the Beke Doline has now been expanded to record flow and dissolution at a high sideslope experiment plot (both overland and at the soil–rock interface), on a group of exposed bedrock pinnacles, and in the underlying cave. It is hoped to communicate the results in a future report.

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